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In situ Cryogenic STEM of Correlated Electronic Materials

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The combination of scanning transmission electron microscopy (STEM) and *in situ* stimuli has enabled a deeper understanding of the behavior of materials and devices [1]. Temperature is a critical parameter to vary *in situ* in order to visualize the nature and behavior of phase transitions such as ferroelectric ordering. In correlated electronic materials, many of the phenomena of interest, including electronic symmetry breaking, occur at low temperatures and thus require stable cryogenic capabilities. Recently, this demand has been met in a handful of atomic-resolution studies of low-temperature phenomena such as charge ordering [2], trimerization [3], ferroelectricity [4,5], and interface superconductivity [6]. Going further, supplementing cryogenic STEM with synergistic methods such as theoretical modelling of phase transitions or *in situ* biasing may deepen our insights into the nature of electronic and structural ordering in correlated materials.

A first application shows how a combination of atomic-resolution cryogenic STEM, picometer lattice tracking, and symmetry-motivated Landau theory reveals the microscopic mechanism underlying charge ordering in a half-doped manganite [7]. In the symmetry-lowering phase transition from the disordered to the ordered state, the order parameter (OP), in this case a periodic lattice distortion with a wavevector $\mathbf{k} = (1/2,0,0)$, is accompanied by secondary OPs with $\mathbf{k} = (1,0,0)$. Landau theory and density functional theory calculations show that the primary and secondary OPs couple in an unusual, non-linear fashion. By selectively mapping these distinct displacement modes using picometer lattice tracking of cryogenic STEM data, we reveal a spatial interplay between the secondary modes and the nature of the charge-ordered ground state, confirming the theoretical predictions.

In a second application of cryogenic STEM, we discuss progress in combining low-temperature imaging capabilities with *in situ* and *ex situ* electrical characterization. Recently, cryogenic holders with electrical feedthroughs have been deployed to heat samples to intermediate cryogenic temperatures using MEMS-based heaters [8,9]. Here, we use an 8+ electrical feedthrough design to both change the temperature and perform 4-probe measurements on quasi-1D charge density wave materials exfoliated on pre-patterned MEMS chips. The charge density wave manifests as anomalies in the resistivity curves and as superlattice peaks in Fourier space. Expanding these *in situ* capabilities promises to reveal and tune the microscopic mechanisms underlying exotic electronic phases [10].

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